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Enhancing VVER Annular Proliferation Resistance Fuel With Minor Actinides

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Absract:

Key aspects of the Global Nuclear Energy Partnership (GNEP) are to significantly advance the science and technology of nuclear energy systems and the Advanced Fuel Cycle (AFC) program. The merits of nuclear energy are the high-density energy, and low environmental impacts i.e. almost zero greenhouse gas emission. Planned efforts involve near-term and intermediate-term improvements in fuel utilization and recycling in current LWR as well as the longer-term development of new nuclear energy systems that offer much improved fuel utilization and proliferation resistance, along with continued advances in operational safety. The challenges are solving the energy needs of the world, protection against nuclear proliferation, the problem of nuclear waste, and the global environmental problem.

To reduce the spent fuel for storage and enhance the proliferation resistance for the intermediate-term, there are two major approaches (a) increase the discharged spent fuel burnup in the advanced LWR (Gen-III Plus), which not only can reduce the spent fuel for storage, but also increase the ^{238}Pu and ^{240}Pu isotopes ratio to enhance the proliferation resistance, (b) use of transuranic nuclides (^{237}Np and ^{241}Am) in the high burnup fuel, which can drastically increase the proliferation resistance isotope ^{238}Pu /Pu ratio.

For future advanced nuclear systems, the minor actinides are viewed more as a resource to be recycled, or transmuted to less hazardous and possibly more useful forms, rather than simply as a waste stream to be disposed of in expensive repository facilities. In this paper, a typical pressurized water reactor (PWR) VVER-1000 annular fuel unit lattice cell model with UO_2 fuel pins will be used to investigate the effectiveness of minor actinide reduction approach (MARA) for enhancing proliferation resistance and improving the fuel cycle performance. We concluded that the concept of MARA, involves the use of transuranic nuclides (^{237}Np and/or ^{241}Am), can not only drastically increase the ^{238}Pu/Pu ratio for proliferation resistance, but also can serve as a burnable absorber to hold-down the initial excess reactivity. It is believed that MARA can play an important role in atoms for peace and the intermediate term of nuclear energy reconnaissance.

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1. INTRODUCTION

Key aspects of the Global Nuclear Energy Partnership (GNEP) are to significantly advance the science and technology of nuclear energy systems and the Advanced Fuel Cycle (AFC) program. GNEP key elements are: (1) a proliferation-resistant process to separate usable elements in spent nuclear; (2) the reduction of plutonium and minor actinides; and (3) an advanced fuel cycle nuclear system. To accomplish these goals, the international cooperation is very important, and public acceptance is crucial. Planned efforts involve near-term and intermediate-term improvements in fuel utilization and recycling in current LWR as well as the longer-term development of new nuclear energy systems that offer much improved fuel utilization and proliferation resistance, along with continued advances in operational safety. It consists of both innovative nuclear reactors and innovative research in separation and transmutation.

The challenges are solving the energy needs of the world, protection against nuclear proliferation, the problem of nuclear waste, and the global environmental problem. The merits of nuclear energy are the high-density energy, and low environmental impacts i.e. almost zero greenhouse gas emission. The new deployment of nuclear energy reconnaissance is the key to a secure economy and environment. Nuclear energy is clearly needed for future solutions. To reduce the spent fuel for storage and enhance the proliferation resistance for the intermediate-term, there are two major approaches (a) increase the discharged spent fuel burnup in the advanced LWR (Gen-III Plus), which not only can reduce the spent fuel for storage, but also increase the ^{238}Pu and ^{240}Pu isotopes ratio to enhance the proliferation resistance, (b) use of transuranic nuclides (^{237}Np and ^{241}Am) in the high burnup fuel, which can drastically increase the proliferation resistance isotope ^{238}Pu /Pu ratio.

2. MINOR ACTINIDES (MA) REDUCTION APPROACH IN LWR

Issues of nuclear waste and proliferation are directly related to the fuel cycle. Worldwide, about 7,000 tons of spent fuels are discharged by nuclear plants. On approaches to transmutation fuel cycle strategies, comparison of recycling of actinides in LWR and in dedicated fast actinides burner reactors indicated that the transmutation through LWR has been found to be more efficient than that through fast reactors in the intermediate term of future nuclear energy. By mixing minor actinides (MA) in the LWR high burnup fuel, which is the Minor Actinides Reduction Approach (MARA),

three major goals can be achieved, (1), effectively reducing the MA storage volume, (2), enhancement of the proliferation resistance, and (3) serving as a burnable absorber to improve the fuel cycle performance.

The overall goal of proliferation resistance is to prevent the extraction of nuclear materials useful for nuclear weapon production from facilities used for civilian nuclear power applications. Based on critical mass considerations, the ^{235}U enrichment limit from proliferation resistance is 20 wt%. However, unlike uranium, any isotopic mix of plutonium has a finite critical mass, i.e., a potential explosive material. Hence, there is no general isotopic concentration threshold for plutonium isotopes from a critical mass point of view. Nevertheless, the suitability for weapons usage varies significantly for plutonium isotopes. Table 1, which is reproduced from Ref. 1, lists the important characteristics of plutonium isotopes. ^{238}Pu, ^{240}Pu, and ^{242}Pu have high spontaneous neutron generation, which reduces the bomb yield significantly. ^{238}Pu also has a high decay heat, which further complicates the design of explosive devices. MARA burning minor actinides of ^{237}Np and/or ^{241}Am in the high burnup fuel can transmute MA and decay to ^{238}Pu in LWR, which is also the subject of Protected Plutonium Production $(P^3)^2$ approach. In the following work, a typical pressurized water reactor (PWR) lattice unit cell model with UO_2 fuel pins will be used to investigate the effectiveness of MARA for enhancing proliferation resistance and improving long fuel cycle performance to satisfy the intermediate-term goal for future nuclear energy systems.

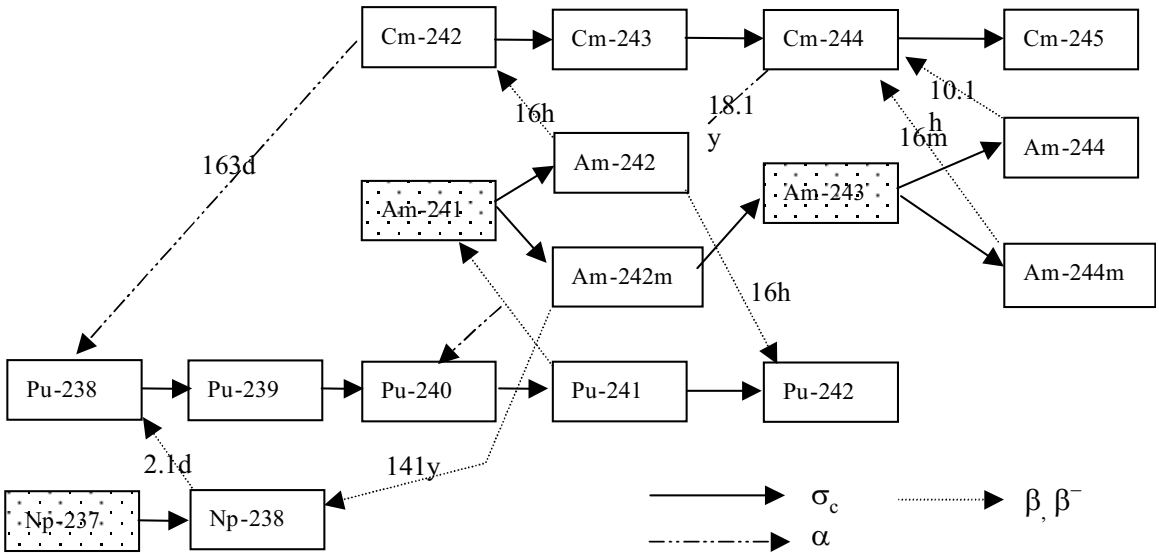
Table 1: Plutonium isotope properties important to proliferation resistance

	Half-life (years)	Spontaneous fission neutrons (n/kg/sec)	Decay heat (Watt/kg)	Bare critical mass (kg)
Pu-238	87.7	2,600,000	560	10
Pu-239	24,100	22	1.9	10
Pu-240	6,560	910,000	6.8	40
Pu-241	14.4	49	4.2	10
Pu-242	376,000	1,700,000	0.1	100

For future advanced nuclear systems, the MAs are viewed more as a resource to be recycled, or transmuted to less hazardous and possibly more useful forms, rather than simply as a waste stream to be disposed of in expensive repository facilities. As a result, they play a much larger part in the design of advanced systems and fuel cycles, not only as additional sources of useful energy, but also as direct contributors to the reactivity control of the systems into which they are incorporated. Figure 1 shows the MA buildup and decay chains that are most commonly considered in the design of advanced reactors and fuel cycles. As shown in Figure 1, ^{237}Np and ^{241}Am can be transmuted and decayed to the highly proliferation resistant isotope ^{238}Pu.

In the following study, a typical pressurized water reactor (PWR) VVER-1000 annular fuel unit lattice cell model with UO_2 fuel pins will be used to investigate the effectiveness of MARA for enhancing proliferation resistance and improving the fuel cycle performance in the intermediate term goal for future nuclear energy systems.

Figure 1. Buildup and decay chains for the MAs. Shaded boxes represent materials with long half-lives that make them of particular interest for transmutation.



3. VVER-1000 UNIT LATTICE CELL MODEL AND MARA STUDY CASES

A typical PWR VVER-1000 unit lattice cell, as shown in Figure 2, has been chosen as the basis for the fuel neutronics analysis. UO_2, Gd_2O_3, NpO_2, and AmO_2 with 95% of theoretical density are used. The fuel rods have a radius of 0.455 cm and are clad with 0.069 cm of Zr. The annular fuel pins are arranged in a hexagonal fuel lattice. The detailed lattice cell parameters are tabulated in Table 2.

Table 2: VVER-1000 unit lattice cell parameters

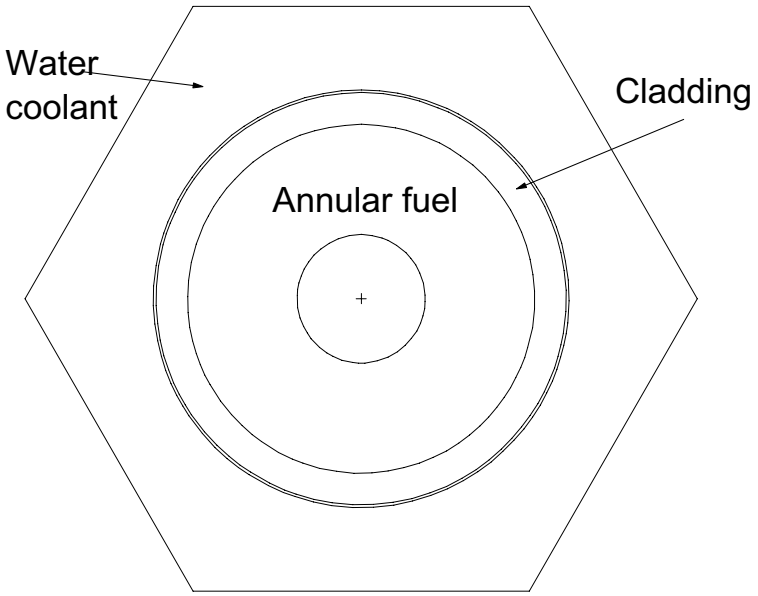
Lattice pitch	1.275 cm
Fuel pin outer radius	0.455 cm
Cladding thickness	0.069 cm
Pellet outer radius	0.38 cm
Pellet central hole radius	0.14 cm

Increasing the fuel discharge burnup can improve the proliferation resistance and reduce the spent fuel storage volume. In this study, UO_2 with ^{235}U enrichment of 4.4 wt% was used. For the high burnup fuel with ^{235}U enrichments of 4.4 wt%, one burnable absorber Gd_2O_3 case, and three mixed oxide (MO) MA cases for UO2+NpO_2, UO_2+NpO_2+AmO_2, and UO_2+AmO_2 were established. The ^{235}U enrichment, Gd_2O_3, NpO_2, and AmO_2 composition of the 5 study cases are summarized in Table 3.

Table 3: UO_2 - ^{235}U enrichment (4.4 wt%), Gd_2O_3, NpO_2, and AmO_2 composition of the 5 study cases

ID	Gd_2O_3 (wt%)	NpO_2 (wt%)	AmO_2 (wt%)
Case-1	--	--	--
Case-2	0.028	--	--
Case-3	--	0.45	--
Case-4	--	--	0.45
Case-5	--	0.2	0.2

Figure 2. Typical VVER lattice unit cell model with a pin pitch of 1.275 cm.



4. MONTE CARLO BURNUP METHOD – MCWO

The physics analyses were performed using the computer code MCNP.[2] In addition, the validated fuel burnup methodology MCNP coupled with ORIGEN2,[3] or MCWO,[4] was used. MCWO has been verified at the Idaho National Laboratory (INL) by benchmarking calculated flux magnitudes with measured flux levels for several experiments and in several test positions of the ATR core.[5,6] Computer codes MCNP, MCWO, and ORIGEN2 are contained in the INL listing of qualified codes.

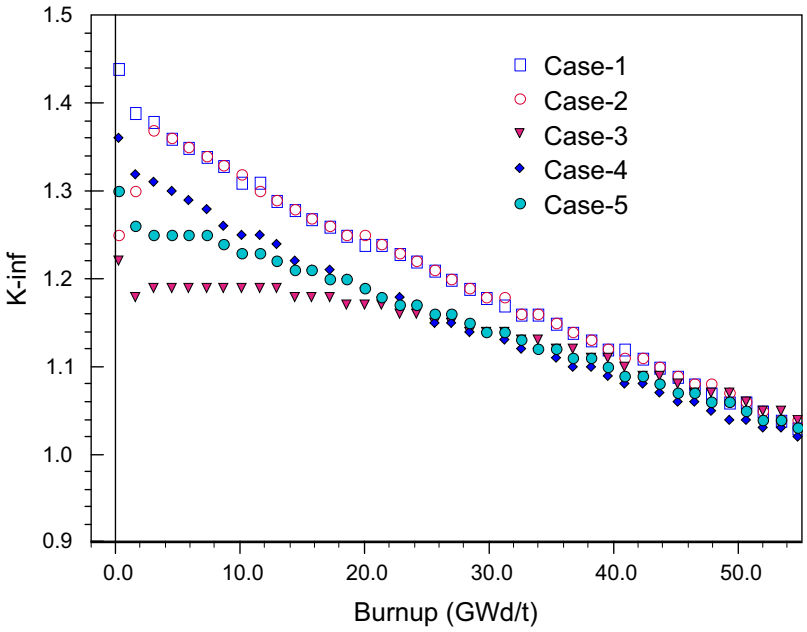
5. RESULTS AND DISCUSSION

MCWO-calculated results for the five case studies will be discussed in the following. The burnup time interval is 1.4 GWd/t. For each time step, an MCNP KCODE

calculation with 2000 source neutrons for 90 cycles is run, requiring ~5 minutes of CPU time on a workstation with two dual-core 2.86 GHz XEON processors. The fission tally calculation can achieve a 1σ standard deviation of 0.2% or less.

The MCWO-calculated K-inf for Cases-1 to -5 versus burnup are plotted in Figure 3. For K-inf = 1.02, Figure 3 shows that the discharged burnup of all five Cases can reach 54 GWd/t. The higher burnup UO_2 fuel with ^{235}U 4.4 wt% can reduce the spent fuel volume proportionally, which benefits the spent fuel storage concerns. From the Figure 3, it clearly shows that MARA mixed fuel can hold down the initial excess reactivity as well as burnable absorber Gd_2O_3. However, due to Gd fast depletion rate, the K-inf will increase to 1.37 at the burnup of 3.08 GWd/t. The best fuel cycle performance is Case-3 with AmO_2 0.45 wt%, which not only can hold down the initial excess reactivity, but also keep the K-inf a very desirable flat profile versus burnup. As a result, the ^{241}Am can serve as a burnable absorber to effectively hold down the initial excess reactivity.

Figure 3. K-inf comparison of VVER-1000 lattice unit cell Cases-1 to -5 versus burnup



One of the criteria in the definition of spent fuel standard, as defined by the National Academy of Sciences[7] is that the isotopic compositions of the discharged fuel should be about the same as the light water reactor UO_2 spent fuel, particularly, the $^{240}Pu/Pu$ ratio should be greater than 24%. For the MCWO-calculated $^{240}Pu/Pu$, $^{238}Pu/Pu$, and $^{239}Pu/Pu$ ratio profiles versus burnup are shown in Figures 4, 5, and 6. The MCWO-calculated $^{240}Pu/Pu$ ratios for Cases-1 and -2 at the discharged burnup (54 GWd/t) can reach about 25.2%, respectively, as shown in Figure 4, which all can meet the spent fuel standard. The MCWO-calculated $^{240}Pu/$ Pu ratios for Cases-3, -4, and -5 at the discharged burnup can reach 22.1%, 21.6, and 22.0%, respectively. Although, the $^{240}Pu/Pu$ ratios are marginal less than 24% for Case-3, -4, and -5, at the discharged burnup, we will discus in the following, that their much better proliferation resistance $^{238}Pu/Pu$ ration are quite

higher than the Case-1 and -2's 2.8% at the discharged burnup. In addition, G. Kessler[9] pointed it out for ^{238}Pu/Pu above 6%, can be considered it's proliferation resistant as effective as 235U < 20% or 233U < 12%. As a result, the (^{238}Pu+^{240}Pu)/Pu ratio is larger than 24% and meets the spent fuel standard.

The MCWO-calculated ^{238}Pu/Pu ratio profiles versus burnup are shown in Figures 5. Figure 5 shows that the fraction of ^{238}Pu in the discharged fuel, increases with burnup, which can much better enhance proliferation resistance. Figure 5 also shows that the fraction of ^{238}Pu in Case-3 drastically increases to about 12.3%, then, levels off at 9.5%, due to the short β-decay time (2.1-day) of ^{238}Np (see Figure 1). For Case-4, the transmutation of the additional ^{241}Am chain, with the long α-decay time of ^{242}Cm, causes the fraction of ^{238}Pu to peak at 14.3% at a burnup of 24 GWd/t, then, decreases to about 14.3%. For Case-5, the transmutation of the ^{237}Np and ^{241}Am chain with the long α-decay time of ^{242}Cm causes the fraction of ^{238}Pu to reach a peak of 12.9% at a burnup of 24 GWd/t, then, decrease to about 11.0%. In summary, Figure 5 shows that the fraction of ^{238}Pu in Cases-3, -4, and -5 increases to a peak of about 12.3, 15.8, and 12.9%, respectively. We conclude that the discharged spent fuel of Cases-4 and -5 can effectively enhance proliferation resistance. The combined fractions of ^{238}Pu and ^{240}Pu can still meet the spent fuel standard. There is a concern that at the low burnup (~10 GWd/t) the ^{240}Pu/ Pu ratios are less than 6.5%, which qualify as weapons-grade Pu. However, due to the short decay time (2.1-day) from ^{238}Np to ^{238}Pu, MARA can provide a high fraction of ^{238}Pu at the low burnup while providing adequate proliferation resistance. For reference, the MCWO-calculated ^{239}Pu/Pu ratio profiles versus burnup are shown in Figures 6, which shows that MARA Cases-3 to -5 have lower 239Pu/Pu ratio (45 atom%) than the UO2 Cases-1 and -2 (49 atom%).

Figure 4. ^{240}Pu/Pu ratio profiles comparison of Cases-1, -2, and -3 versus burnup

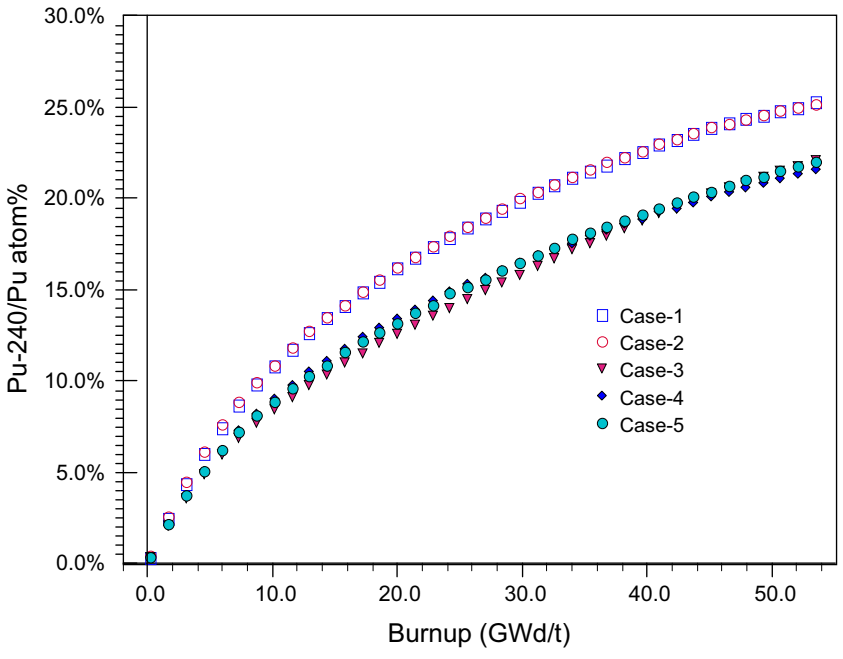


Figure 5. ^{238}Pu/Pu ratio profiles comparison of Cases-1, -2, and -3 versus burnup

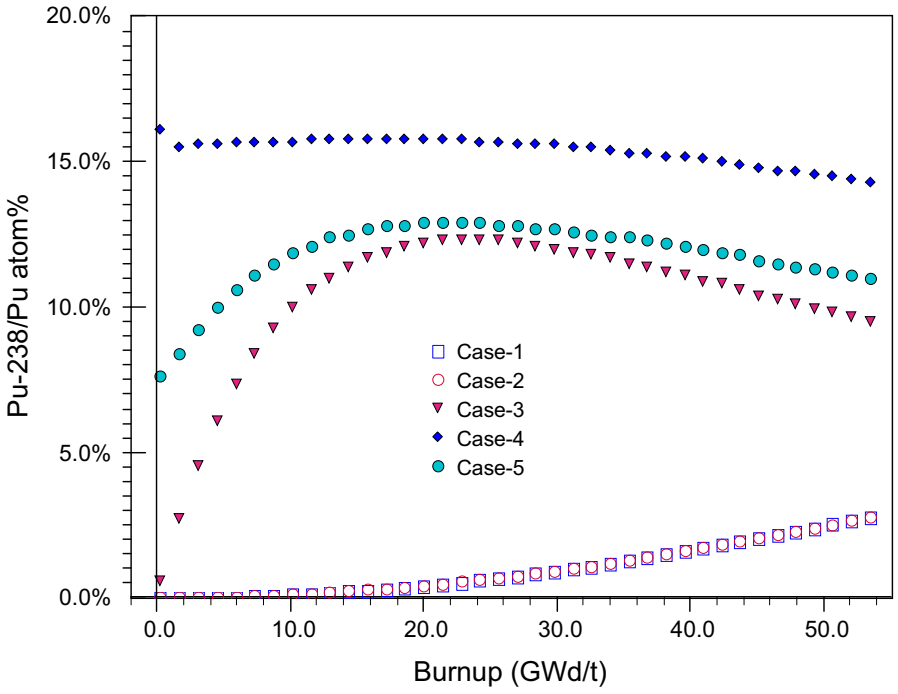
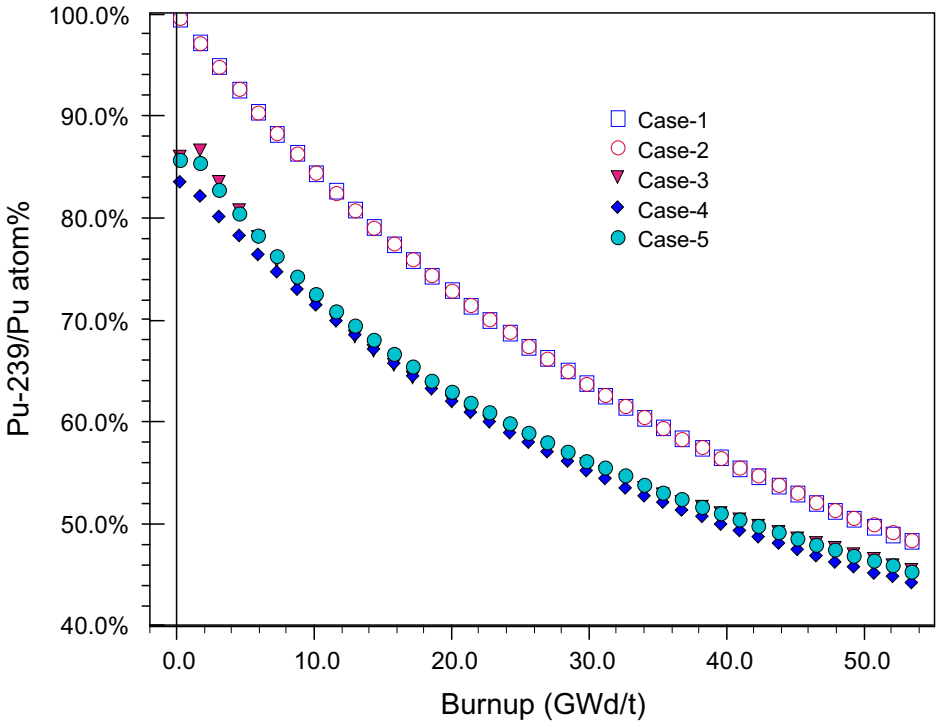


Figure 6. ^{239}Pu/Pu ratio profiles comparison of Cases-1 to -5 versus burnup



6. CONCLUSIONS

Based on the studies presented herein, it is strongly believed that the concept of MARA, involves the use of transuranic nuclides (^{237}Np and/or ^{241}Am), can not only drastically increase the ^{238}Pu/Pu ratio for proliferation resistance, but also can serve as a burnable absorber to hold-down the initial excess reactivity. It is believed that MARA can play an important role in atoms for peace and the intermediate term of nuclear energy reconnaissance.

The known disadvantages of MARA are:

1. ^{237}Np and ^{241}Am in the fuel will generate more ^{4}He, which will affect the fission gas release performance.
2. The need to have improved transuranic cross-section library data.
3. The ^{237}Np is a controlled nuclear sensitive material.

And, the encouraging advantages of MARA are:

1. The high burnup of the spent fuel (^{235}U 4.4-wt %) is well developed and becoming a standard for LWR fuel reloading, which can effectively reduce the spent fuel storage volume.
2. ^{237}Np and/or ^{241}Am can drastically increase the fraction of ^{238}Pu to enhance the proliferation resistance.
3. ^{241}Am not only can increase the fraction of ^{238}Pu, but also can be used as a burnable absorber to reduce the initial excess reactivity.
4. ^{237}Np can be transmuted quickly with β-decay (half-life 2.1 days) to a high fraction of ^{238}Pu, which can effectively enhance the proliferation resistance for early removal of low burnup fuel.

As a result of these concerns, it is believed that Case-4 ^{235}U 4.4-wt% with ^{241}Am 0.45-wt% is the best candidate MARA fuel, which not only can achieve the high burnup design goal, but also can achieve the proliferation resistance enhancement goal.

7. REFERENCES

1. J. C. Mark, "Explosive Properties of Reactor-Grade Plutonium," *Science & Global Security*, **4**, 111-128, 1993.
2. M. SAITO, "Advanced Core Concepts with Enhanced Proliferation Resistance by Transmutation of Minor Actinides," Proceedings of GLOBAL 2005 Tsukuba, Japan, Oct 9-13, 2005 Paper No. 172.
3. J. F. Briesmeister (Editor), "MCNP—A General Monte Carlo N-Particle Transport Code, Version 4C," LA-13709, Los Alamos National Laboratory (2000).
4. A. G. Croff, "ORIGEN2: A Versatile Computer Code for Calculating the Nuclide Compositions and Characteristics of Nuclear Materials," *Nuclear Technology*, Vol. 62, pp. 335-352, 1983.
5. G. S. Chang and J. M. Ryskamp, "Depletion Analysis of Mixed Oxide Fuel Pins in Light Water Reactors and the Advanced Test Reactor," Nucl. Technol., Vol. 129, No. 3, p. 326-337 (2000).
6. F. W. Ingram, R. G. Ambrosek, G. S. Chang, and D. J. Utterbeck, "JAPEIC Irradiation Report for the SFT-1, SFT-3 and FELI-2/3 Specimens," August 2001.
7. G. S. Chang, "ATR WG-MOX Fuel Pellet Burnup Measurement by MONTE CARLO – MASS Spectrometric Method," *Proceedings of the International Conference on the New Frontiers of Nuclear Technology: Reactor Physics, Safety and High-Performance Computing, ANS 2002 RPD Topical Meeting, Seoul, Korea*, October 7-10, 2002.
8. National Academy of Sciences Committee on International Security and Arms, Management and Disposition of Excess Weapons Plutonium, National Academy Press, 1994.
9. G. Kessler, "Plutonium Denaturing by 238Pu," Nucl. Sci. & Eng.: **155**, 53-73 (2007).